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DISTRIBUTION OF IRON, MANGANESE, ZINC AND ATRAZINE IN GROUNDWATER IN PARTS OF PALAR AND CHEYYAR RIVER BASINS, SOUTH INDIA

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Abstract. A study was carried out in a part of Palar and Cheyyar river basin to evaluate the current status of iron, manganese, zinc and atrazine concentrations, their origin and distribution in groundwater. Groundwater samples were collected during post-monsoon (March 1998 and February 1999) and pre-monsoon (June 1999) periods from 41 sampling wells distributed throughout the study area. The groundwater samples were analyzed for trace metals using AAS and atrazine using HPLC. The concentration of the trace elements in groundwater is predominant during pre-monsoon period. Distribution pattern indicates that the concentration of these elements increases from west to northeast and towards Palar river. Lower concentrations in the central part may be due to recharge of fresh water from the lakes located here. During most of the months, as there is no flow in Palar river, the concentrations of trace elements in groundwater are high. Drinking water standards indicate that Mn and Zn cross the permissible limit recommended by EPA during the pre-monsoon period. A comparison of groundwater data with trace element chemistry of rock samples shows the abundance of trace elements both in the rock and water in the order of Fe > Mn > Zn and Fe > Zn > Mn. This indicates that iron in groundwater is derived from lithogenic origin. Further, Fe, Mn and Zn have good correlation in rock samples, while it is reverse in the case of water samples, indicating the non-lithogenic origin of Mn and Zn. Atrazine (a herbicide) was not detected in any of the groundwater samples in the study area, perhaps due to low-application rate and adsorption in the soil materials.

Keywords: atrazine, groundwater, iron, manganese, zinc, Palar basin, India, Tamil Nadu

1. Introduction

Trace metal concentrations in groundwater have attracted the attention of researchers worldwide for a variety of reasons such as (a) their significance related to public health problems, (b) their need in relation with plant growth and (c) academic interest to understand the mechanism of metal transport in aqueous environment. The contribution of man-made activities to major ion composition in groundwater is limited in comparison to trace metals. Trace metals are contributed by agricultural activities and by rock weathering. Pawar and Nikumbh (1999) studied the trace element geochemistry of groundwater from Behedi Basin, Maharashtra, and reported that agricultural activities (micronutrient fertilizers) contributed trace elements to the groundwater. Further, they concluded that most of the trace elements entered the aquifer during rainy season. Edmunds *et al.* (1992) studied the trace metals in interstitial waters from sandstones in southern Britain and suggested that dissolution of primary minerals is the source of trace metals in the interstitial waters of the unsaturated zone. Encounter with anoxic water is the reason for elevated Fe and Mn concentrations in groundwater (Kraft *et al.*, 1999). Romic and Romic (2003) studied the heavy metal distribution in agricultural topsoils in an urban area, Croatian, Zagreb and reported that Fe and Mn are mainly from geogenic (pedogenic origin) and Zn from atmospheric deposition and anthropogenic influence. Nriagu and Pacyna (1988) believe that the mobilization of heavy metals in the biosphere, caused by human activity, significantly increased the circulation of toxic metals through soil, water and air. Khan *et al.* (1996) studied the mobility of heavy metals in soil and reported that the application of chemical fertilizers such as urea, ammonium phosphate, ammonium sulphate and potassium chloride in soils enhance the mobility of Ni, Mn and Cr to a considerable extent.

In addition to trace metals, groundwater contamination by pesticides has become a major issue in the world. Conservation tillage, which generally requires the application of herbicides, has increased tremendously. With reduced runoff and increased infiltration, conservation tillage may increase the leaching of herbicides to groundwater. Atrazine is the most widely used herbicide. It has been probably detected more widely in groundwater than any other herbicides. Junk *et al.* (1980) measured atrazine concentrations as high as 88 ppb in groundwater samples in Nebraska. Peak concentration was observed at the end of the irrigation season in shallow wells down-gradient from irrigated fields. The vertical and areal distributions of atrazine were closely associated with those of nitrate-nitrogen. Spalding *et al.* (1980) also reported finding atrazine in the water under irrigation fields in Merrick County, Nebraska.

The present study was carried out to determine the current status of Fe, Mn, Zn and atrazine concentrations, their origin and distribution in the groundwater in parts of Palar and Cheyyar river basins, South India. In late 1980s, the Institute of Water Studies, Public Works Department (PWD), Government of Tamil Nadu, made an assessment of the water resources and management strategies for this region of Palar basin. The PWD (2000) explored the water resources scenario and found that there was no surplus surface water resource in this area and hence groundwater had to be conserved for various uses. Rajmohan *et al.* (2000) studied the major ion chemistry of groundwater in Kancheepuram region, a part of Palar basin and concluded that silicate weathering reaction is the probable source of sodium, calcium and magnesium in groundwater in this region. Thus, no earlier investigation of trace element chemistry and pesticide concentration in groundwater has been attempted in this area.

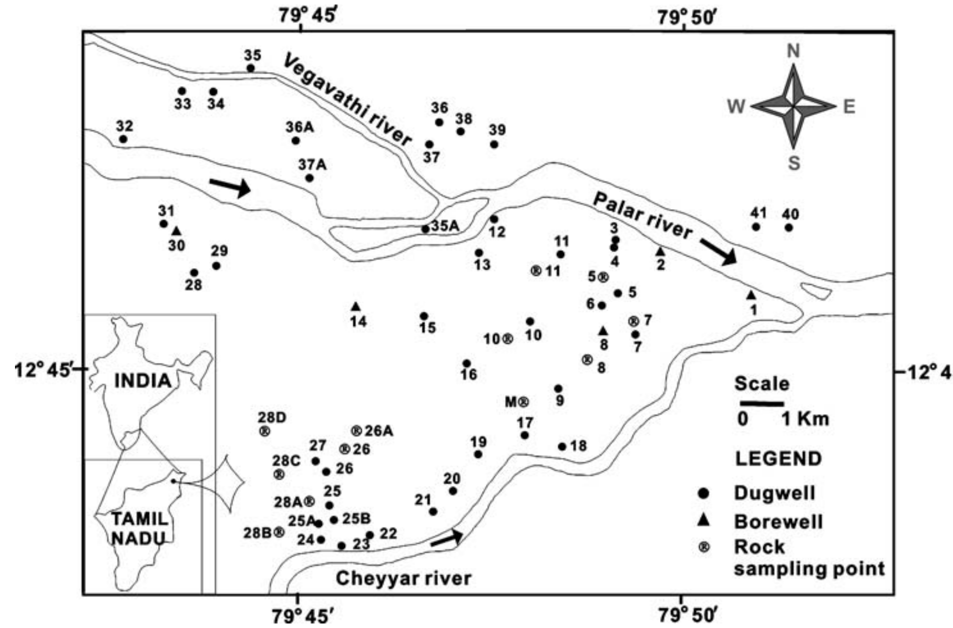

Figure 1. Location of the sampling stations in the study area.

2. Description of the Study Area

The study area (Figure 1) is situated in Kancheepuram District in Tamil Nadu State, India. It forms a part of the Palar and Cheyyar river basins, and is located 70 km west of Chennai city, India. It covers an area of 234 km^2 extending approximately 18 km along north and south and about 13 km along east and west. The maximum elevation is 94 m above mean sea level in the eastern side of the study area; the minimum elevation is about 50 m. Generally, the area slopes towards east. The Vegavathi river flows in the northwestern part of the study area.

2.1. CLIMATE AND RAINFALL

The study area has dry climatic condition with the maximum temperature of around 39 °C during the months of April and May, and minimum temperature of around 21 °C during the months of November and December. It receives an average annual rainfall of 1113 mm, of which 60% is contributed by the NE (north–east) monsoon, from October to December; the rest is during the SW (south–west) monsoon, from June to September. The SW monsoon sets in the month of June and brings some rain until the end of September. The NE monsoon sets in the month of October, which brings heavier rains till December. Heavy rains are often associated with

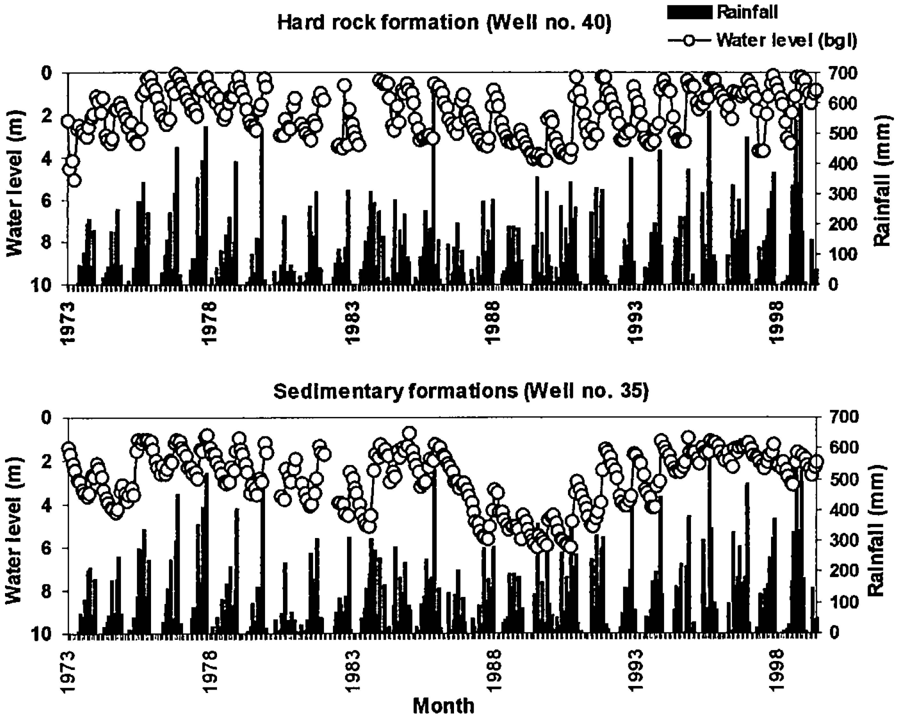

Figure 2. Relation between water level fluctuation and rainfall variation (1973–1999).

depressions and storms, which generally occur in the Bay of Bengal during the NE monsoon. Highest rainfall was received during 1998 as per the last 20 years record (Figure 2). Rainfall is the major source for groundwater recharge. The rivers in this region flow only for a few days in a year after heavy rains during monsoon. The study area has number of lakes, which are also the major source for recharge.

2.2. GEOLOGY

The study area is composed of sedimentary and crystalline formations (Figure 3). They are unconformably overlain by sandy and clayey soils of recent to sub-recent age. The sedimentary rock types include sandstone and shale. The crystalline formations are charnockite, granitic gneiss and ultra basic rock. The crystalline charnockite and granitic gneiss of Archean age have been intruded by amphibolites, dykes of dolerite and occasionally by veins of quartz and pegmatites. The gneisses of this area have quartz, feldspars (potash feldspars and albite), hornblende, biotite, etc. The acid charnockite of this area has quartz, K-feldspars, hypersthene and biotite minerals of coarse-grained nature. Ultrabasic rocks include small patches

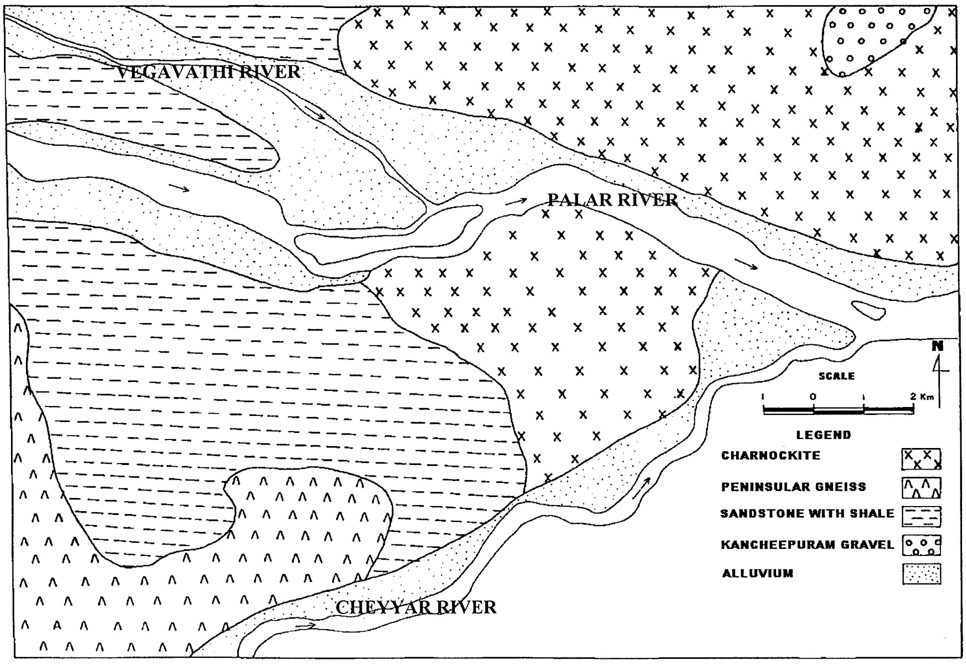

Figure 3. Geology map of the study area.

of maficgranulites and pyroxenite occurring within charnockites. Alluvial deposits constitute the youngest formation consisting of sands and clays, and occurring along river courses.

2.3. HYDROGEOLOGY

Groundwater occurs under water table conditions in weathered and fractured portions of crystalline rocks. The pore spaces developed in the weathered mantle act as shallow granular aquifer and form potential water-bearing zones. The depth of wells in this region is up to 10 m. Most of the wells in the hard rock region are large diameter dug wells. Generally, the water level in the crystalline formations is between 6 and 9 m below ground level. Sometimes, during summer, these wells are completely dry. Alluvium is the most important sedimentary formation, occurring on both sides of the river. The alluvium is essentially composed of sand with intercalated clay. In these formations, wells have been dug up to a depth of 23 m. Most of the wells are bore and dug cum borewells. The water level in these wells fluctuates between 5 and 20 m below ground level.

2.4. IRRIGATION PRACTICES

The study area is intensively cultivated, with three crops in a year. Mostly paddy is cultivated. Sometimes, sugarcane is cultivated, which is a 12-month crop. The main cropping season is from September to January, when paddy is grown. During this period, both river water and groundwater are used for irrigation. The second cropping season is from February to May when paddy, vegetables, pulses and groundnut are grown. These crops are mainly dependent on tank water and groundwater. Tank water is used for irrigation during October–May. Paddy and other crops are sometimes grown in some parts of the area from May to September, mainly dependent on groundwater. The more commonly used fertilizers for paddy in the study area are urea, NPK complex, muriate of potash and zinc sulphate. Atrazine was used as herbicide in sugarcane fields.

3. Materials and Methods

3.1. GROUNDWATER SAMPLING AND ANALYSIS

Groundwater samples were collected during post-monsoon (March 1998 and February 1999) and pre-monsoon (June 1999) periods from 41 sampling wells (Figure 1). Water samples were collected in clean polythene bottles. All sampling bottles were soaked with 1:1 HNO_3 and washed using detergent. These bottles were then rinsed with double distilled water. At the time of sampling, sampling bottles were thoroughly rinsed three times using the groundwater to be sampled. In the case of bore wells, the water samples were collected after pumping the water at least for 15 min. In the case of open wells, water samples were collected 30 cm below the water level using depth sampler.

Samples collected were transported to the laboratory on the same day and they were filtered using 0.45-mm Millipore filter paper and acidified with nitric acid (Ultra pure, Merck) for trace metal analyses. The concentrations of Zn, Fe and Mn ions were measured using atomic absorption spectrometer (Varion Spectra 200) by direct aspiration (APHA, 1989). While performing the analysis, two sets of internal standards were run, one at the beginning and other in between the analyses, to have a check on the accuracy and precision of the results. The detection limits are around 1 μg/L.

For the analysis of atrazine (herbicide), groundwater samples were collected at selected sites from the study area during June 1998 and filtered immediately using 0.45 μm filter paper. Atrazine in the groundwater samples was extracted using C_{18} Sep-Pak® cartridge (Waters Associates, Milford, Massachusetts, U.S.A.) (Ritter *et al.*, 1994). C_{18} Sep-Pak cartridges were preconditioned with 6 mL methanol (HPLC grade) and 6 mL deionized water without letting the cartridge dry out. Then, 250 mL of the filtered sample was loaded into the preconditioned C_{18} Sep-Pak cartridge at a flow rate of approximately 15 mL/min. The cartridge was washed with deionized water (3 mL) and vacuum dried for 15–30 min using a vacuum pump.

Atrazine was then eluted from the cartridge using 4 mL of methanol at a flow rate of 1 ml/min. Eluted samples were transferred to glass vials and volumes were recorded at every stage to determine the concentration factor. To determine the recovery of the procedure, spiked samples in deionized water with known concentrations of atrazine were prepared. Identification and quantification of atrazine was made using HPLC. The elutes were chromatographed using a 25 cm, 5 μm C-18 column (Sperisorb ODS2) that was maintained at 60 °C on a Waters HPLC system (Waters model 600E system controller and pump, model 717 + autosampler, and model 996 photodiode array detector controlled by Millennium 2010, Version 2.00 software). The initial solvent conditions were 20:80 methanol:water (volume:volume) at a flow rate of 1.4 mL/min. Data was collected from the photodiode array at a rate of 1.0 spectrum/s with a resolution of 1.2 nm from 215 to 300 nm. The wavelength selected for quantification was 216 nm. The quantification was made based on retention times, in comparison with corresponding peak height of standards. Elution, identification and quantification were carried out in Deakin University, Australia. In addition to water analysis, some rock samples collected near groundwater sampling wells in the study area were analyzed. Rock samples were analyzed for total Fe, Mn and Zn, as per the procedure given in Shapiro (1975). To carry out detailed studies, apart from collection and analysis of water samples during this study, rainfall data pertaining to the study area were collected from the Public Works Department (PWD).

4. Results and Discussion

Iron, manganese, copper, zinc, lead, silver, etc. are the important major and trace elements that occur in the crystal structure of minerals found in rocks. However, in groundwater, all of them occur as trace elements. Their distribution and occurrence in groundwater, however, depend on the degree of weathering and mobility of these elements during weathering (Handa, 1986). As agricultural activities are very common in the study area, iron, manganese and zinc ions are likely to get leached into the groundwater from micronutrient fertilizers (Pawar and Nikumbh, 1999). Hence, iron, manganese and zinc concentrations in groundwater of the study area were studied.

4.1. IRON

Iron concentration in groundwater of this area generally varies from 8 to 175 μg/L, except well no. 10 (Table I). The average concentration varies from 39 μg/L

TABLE I
Trace element concentrations in groundwater samples

	Post-monsoon (March 1998)			Post-monsoon (February 1999)			Pre-monsoon (June 1999)		
Well no.	Fe (μg/L)	Mn (μg/L)	Zn (μg/L)	Fe (μg/L)	Mn (μg/L)	Zn (μg/L)	Fe (μg/L)	Mn (μg/L)	Zn (μg/L)
1	82	169	176	52	229	80	110	322	221
2	67	8	4	NA	3	8	NA	NA	NA
3	65	12	2	54	25	28	119	156	183
4	67	11	24	16	11	29	120	169	184
5	52	7	1	38	7	34	113	131	194
6	63	9	7	38	9	43	80	120	300
7	44	5	9	57	8	35	72	262	284
8	16	4	47	69	30	102	124	229	275
9	27	4	BDL	41	10	39	155	190	286
10	100	12	3	250	43	37	168	211	295
11	24	6	14	96	11	48	142	115	299
12	16	5	2	138	10	21	143	184	264
13	16	6	3	74	10	42	155	228	247
14	45	6	10	29	6	38	175	207	249
15	28	9	10	63	11	49	166	197	259
16	11	5	BDL	37	8	32	67	142	298
17	39	90	13	76	96	65	101	182	313
18	41	10	8	50	11	59	81	168	334
19	30	17	1	45	BDL	25	92	78	368
20	30	8	8	37	10	35	99	150	379
21	21	9	4	42	26	44	110	91	395
22	32	12	18	27	9	39	19	136	140
23	30	14	17	31	6	30	30	110	152
24	43	13	13	85	10	33	40	148	138
25	39	14	10	42	12	39	45	147	169
26	30	13	13	41	12	27	48	106	178
27	33	18	6	38	13	25	56	66	101
28	36	19	10	84	15	44	68	116	135
29	20	19	24	8	13	27	77	99	164
30	37	14	12	42	15	48	94	57	147
31	38	18	18	39	14	34	87	117	122
32	29	18	33	51	15	39	113	190	149
33	40	19	196	42	15	41	112	210	164
34	38	52	24	60	17	37	114	135	155
35	42	20	9	78	17	35	125	121	130

(*Continued on next page*)

TABLE I
(*Continued*)

	Post-monsoon (March 1998)			Post-monsoon (February 1999)			Pre-monsoon (June 1999)		
Well no.	Fe (μg/L)	Mn (μg/L)	Zn (μg/L)	Fe (μg/L)	Mn (μg/L)	Zn (μg/L)	Fe (μg/L)	Mn (μg/L)	Zn (μg/L)
36	35	19	10	56	14	37	139	140	137
37	30	18	7	58	17	34	140	204	177
38	32	74	18	35	19	30	113	82	140
39	28	19	27	64	16	34	NA	NA	NA
40	31	17	BDL	62	17	30	98	179	125
41	52	25	26	72	16	42	10	50	41
Min.	11	4	1	8	1	8	10	50	41
Max.	100	169	196	250	229	102	175	322	395
Mean	38.51	20.66	21.97	57.93	21.40	38.95	100.51	152.44	212.59
S.D.	18.17	29.27	39.35	39.52	36.46	15.62	46.07	62.19	95.62

BDL – Below detection limit. Min. – Minimum. Max. – Maximum. S.D. – Standard deviation.

(March 1998) to 101 μg/L (June 1999). Samples collected from well no. 10 have high concentration of iron during post-monsoon, which might be due to weathering of laterite. Iron in the groundwater is mostly in the form of inorganic complexes derived from laterite and other types of soils as reported by Ramesh *et al.* (1995). Seasonal variation indicates that the high concentration observed during pre-monsoon (June 1999) might be due to flushing/dissolution of lithogenic and non-lithogenic materials by infiltrating water. In general, rock water interaction and agricultural activities are the major source for high concentration of iron in the study area.

4.2. MANGANESE

The concentration of manganese in groundwater of the study area varies from below detection limit (BDL) to 262 μg/L, with extreme concentrations of 322 μg/L (Well no. 1) (Table I). The average concentration varies from 21 μg/L (March 1998) to 153 μg/L (June 1999). Manganese concentration in groundwater samples of the study area is generally less than 25 μg/L during post-monsoon. Manganese tends to be adsorbed on to the clay, organic matter, freshly precipitated hydrated iron oxides, aluminates, silicates and calcite (Mehrotra and Mehrotra, 2000). During pre-monsoon, however, concentration of manganese varies from 25 to 262 μg/L. Seasonal trend indicates that high concentration observed during pre-monsoon (June 99) seems to be due to flushing or dissolution of non-lithogenic material by infiltrating water.

4.3. ZINC

Zinc belongs to a group of trace metals that are potentially very dangerous for the biosphere. The main sources of pollution are industries and the use of liquid manure, composted materials and agrochemicals such as fertilizers and pesticides in agriculture (Singh, 1994; Romic and Romic, 2003). The concentration of zinc in groundwater of the study area varies from below detection limit (BDL) to 395 μg/L (Table I). During post-monsoon (March 98 and Feb. 99), it varies from BDL to 75 μg/L. However, during pre-monsoon (June 99), it varies from BDL to 395 μg/L. The average concentration varies from 22 (March 98) to 213 μg/L (June 99). Seasonal variation indicates that high concentration is observed during pre-monsoon (June 99). Similar trend was observed in the other elements also. Agricultural activity (fertilizers and micronutrient fertilizer) is the major source for high concentration of zinc in the groundwater of the study area.

To get an idea about the relative abundance of trace elements in the groundwater of the study area, the data were plotted in Figure 4B. It is observed that iron is predominant during post-monsoon, whereas Zn and Mn are predominant during pre-monsoon. As already mentioned, pre-monsoon results show high concentration of all these elements compared to post-monsoon. The southwest monsoon sets in during May-June, which flushes the weathered and non-lithogenic materials (Figure 5). This is the reason for high concentration of all these elements during pre-monsoon.

4.4. DISTRIBUTION OF TRACE ELEMENTS

The distribution of Fe, Mn and Zn in the study area is shown in Figures 6–8, respectively. Distribution pattern indicates that the concentration of trace elements in groundwater of the study area increases from west to northeast and towards the Palar river. High concentrations are observed along the riverbank. Further, these concentrations increase from the central part of this region towards the riverbank. Lower concentrations in the central part may be due to recharge of fresh water from the lakes located here. During most of the months, as there is no flow in Palar river, the concentrations of ions in groundwater are high. Hence, ionic concentrations are high throughout the year, except during the months of November and December. During these months, heavy flow of water in this river dilutes the ionic concentration of groundwater. In addition, topography and groundwater flow pattern are also related to the distribution pattern of the trace elements.

4.5. COMPARISON WITH DRINKING WATER STANDARDS

The maximum allowable concentration of iron in drinking water is 1.0 mg/L, according to WHO report. According to Indian Standard Institute (ISI), the

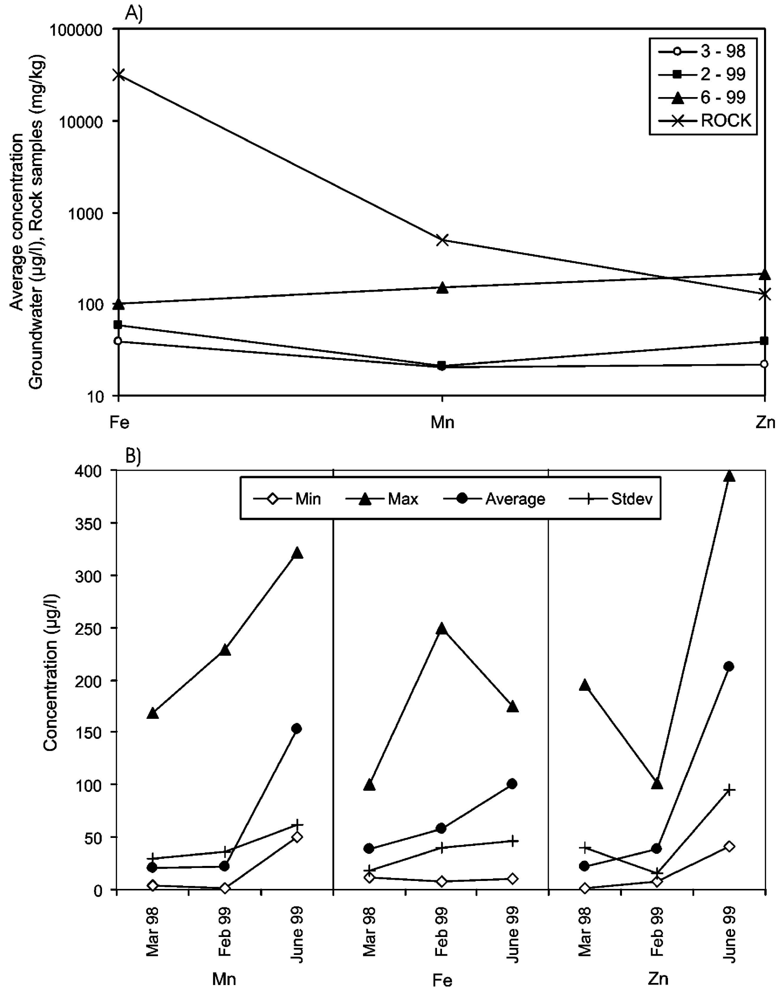

Figure 4. Comparison of Zn, Mn, Fe concentrations in groundwater. Plot A – Average concentration of trace elements in groundwater and rock samples. Plot B – Statistical summary of the trace elements concentrations in groundwater.

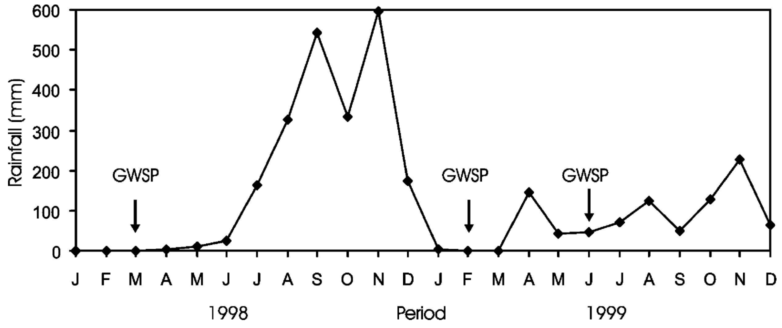

Figure 5. Monthly rainfall variation in the study area. GWSP – Groundwater sampling period.

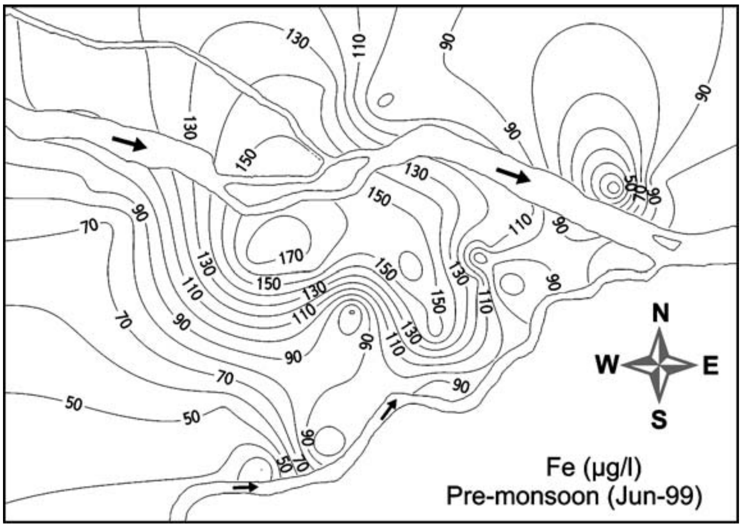

Figure 6. Spatial variation of iron concentration in groundwater.

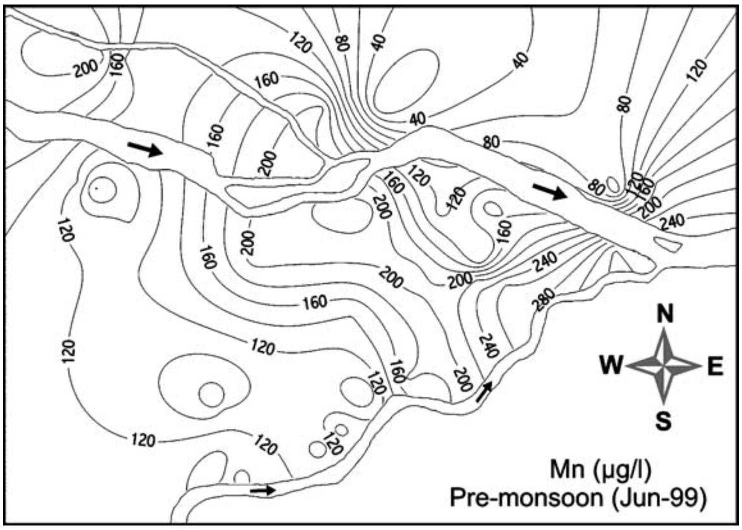

Figure 7. Spatial variation of manganese concentration in groundwater.

permissible concentration of iron in drinking water is 0.3 mg/L. The average concentration of iron is less than 100 μg/L. All samples are within the permissible limit. The maximum allowable concentration and the permissible concentration of manganese in drinking water is 0.5 and 0.1 mg/L, respectively, according to WHO and ISI. The samples collected during post-monsoon (March 98 and February 99) are within the permissible limit. The sample collected during pre-monsoon (June 99),

TABLE II
Trace element concentrations in the rock samples

Well no.	Rock types	Fe (%)	Mn (mg/Kg)	Zn (mg/Kg)
M	Gneiss	8.5201	1519	255.3
5	Gneiss	1.9508	277	161.6
7	Gneiss	2.9359	478	140.5
8	Gneiss	1.8957	273	98.6
10	Gneiss	1.6422	382	112.2
11	Gneiss	2.3366	360	101.4
26	Charnockite	2.8067	373	106.9
26A	Charnockite	2.9347	422	112.7
28A	Charnockite	2.1795	307	118.5
28B	Charnockite	3.6447	583	117.6
28C	Charnockite	2.9687	427	89.3
28D	Charnockite	3.3811	534	127.1

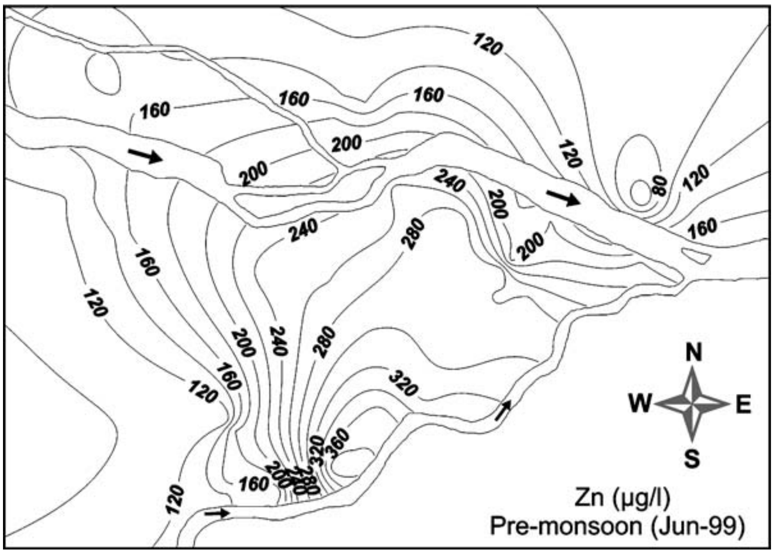

Figure 8. Spatial variation of zinc concentration in groundwater.

however, exceeds the permissible limits of EPA (2002) and ISI standards (50 and 100 μg/L). The maximum allowable concentration and the permissible concentration of zinc in drinking water are 10 and 5 mg/L, respectively, according to ISI. The concentration of Zinc is within the permissible limit during post-monsoon period. During pre-monsoon, however, around 50% of the samples cross the permissible limits of EPA (150 μg/L).

4.6. COMPARISON BETWEEN ROCK AND GROUNDWATER CHEMISTRY

Available geochemical data of the rock samples collected near the sampling wells in the study area show that the elements with high concentrations in the rocks are consistent with those in groundwater only in the case of Fe. The abundance of trace elements in the rock is in the order of Fe > Mn > Zn (Table II, Figure 4A). In the case of groundwater, the order of abundance for the same elements is Fe > Zn > Mn during post-monsoon (March 98 and February 99). This is comparable in the case of Fe concentration. But, during pre-monsoon, the order is reverse (Zn > Mn > Fe). This indicates that the probable source of iron in the groundwater

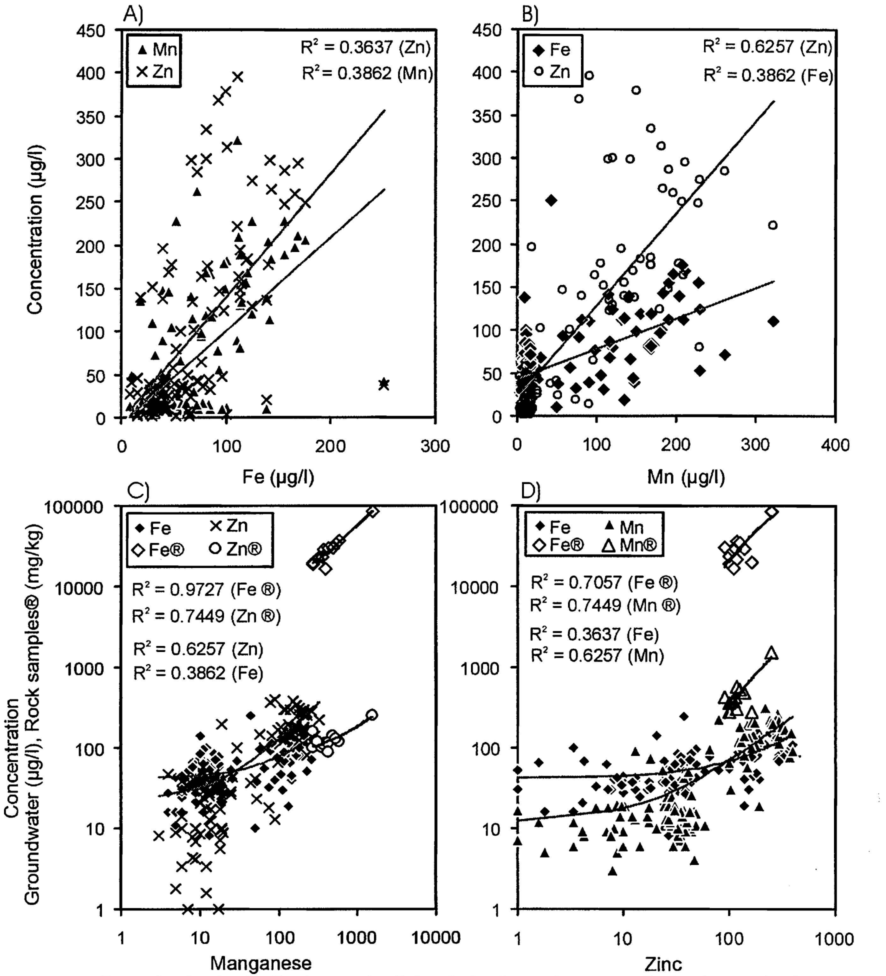

Figure 9. Graphical representation of Fe, Mn and Zn in groundwater and rock samples. Plot A and B – Groundwater samples. Plot C and D – Both groundwater and rock samples.

is mainly lithogenic. The pre-monsoon data, however, indicates the influence of non-lithogenic sources, particularly for Mn and Zn.

The graphical interpretation of trace elements data of both groundwater and rock samples was carried out. Both groundwater and rock data were plotted on the same graph to explain inter elemental relationships (Figure 9). The correlation value (r^2) of rock data indicates that the order is (Fe vs. Mn ($r^2 = 0.97$)) > (Zn vs. Mn ($r^2 = 0.74$)) > (Fe vs. Zn ($r^2 = 0.71$)). In the case of water, the order is (Zn vs. Mn ($r^2 = 0.63$)) > (Fe vs. Mn ($r^2 = 0.39$)) > (Fe vs. Zn ($r^2 = 0.36$)). The graphical plot shows that poor correlation is observed between Fe, Zn and Mn in the groundwater, indicating the non-lithogenic origin of these elements.

4.7. ATRAZINE (HERBICIDE)

Groundwater samples collected from 20 wells in this area were analyzed for atrazine concentration. Atrazine has been widely used as an herbicide for sugarcane in this area. However, atrazine was not detected in any of the groundwater samples, which may be because of low application rate of this herbicide and its adsorption by the soil in this area.

5. Conclusion

A study was carried out in parts of Palar and Cheyyar river basins to evaluate the current status of iron, manganese, zinc and atrazine concentrations, and their origin and distribution. The concentrations of all these trace elements are predominant during pre-monsoon period, which might be due to flushing of weathered materials and non-lithogenic materials during rainwater recharge. Spatial distribution pattern indicates that the concentration of trace elements in groundwater of the study area increases from west to northeast and towards Palar river. Lower concentrations in the central part may be due to recharge of fresh water from the lakes located here. During most of the months, as there is no flow in Palar river, the concentrations of ions in groundwater are high. Mn and Zn concentrations in groundwater are more than the permissible limit of EPA during pre-monsoon period. Trace element concentrations of groundwater were compared with rock samples data. It indicates that the probable source of iron is lithogenic, whereas that of Zn and Mn is non-lithogenic. Further, Fe, Mn and Zn have good correlation in rock samples, while it is reverse in the case of water samples. Atrazine (a herbicide) was not detected in any of the groundwater samples in the study area, which may be due to low-application rate, and adsorption in the soil materials.

The dataset, which is the first of its kind in the area, will serve as reference material in the area for monitoring the vulnerability of this aquifer to pollution in future as a result of intensive agricultural activities.

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